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Density Changes in Plutonium Observed from Accelerated Aging using Pu-238 Enrichment

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Plutonium, because of its radioactive nature, ages from the “inside out” by means of self-irradiation damage and thus produces Frankel-type defects (vacancies and self-interstitial atoms) and defect clusters. The self-irradiation damage in Plutonium-239 occurs mainly by α -particle decay, where most of the damage comes from the U-235 recoil nucleus. The defects resulting from the residual lattice damage and helium in-growth could result in microstructural and physical property changes. Because these self-irradiation effects would normally require decades to measure, with a fraction (7.5 wt%) of Pu-238 is added to the reference plutonium alloy thus accelerating the aging process by approximately 18 times the normal rate. By monitoring the properties of the Pu-238 spiked alloy over a period of about 3.5 years, the properties of plutonium in storage can be projected for periods up to about 60 years. This paper presents density and volume changes observed from the immersion density and dilatometry measurements equivalent to aging the reference plutonium alloys to nine years.

Figure 1 shows the design of a dilatometry system set up to monitor long term growth resulting from the lattice damage and helium in-growth in Pu-238 spiked alloys. Each dilatometer unit consists of a small vacuum controlled-atmosphere sample chamber fitted with three linear variable differential transducers (LVDTs). An LVDT measures minute changes, 1 micron or less, in the position of a push-rod by monitoring changes in the inductance of a detector coil. In the current design, the detector coil is placed outside of the sample chamber. Two different lengths (2 and 3 cm) of alloy specimens are used to differentiate between surface oxidation and volumetric swelling in the materials. These specimens are place in the copper well located inside the dilatometer system. A reference low expansion glass (Zerodur) is also place in the copper well to monitor the stability of the dilatometry system.

Figure 2 shows a possible volume change behavior of plutonium as a function of time. Our goal is to perform continuous measurement of volume expansion in plutonium alloys caused by self-irradiation damage.

During the early stage of measurement, samples at 35, 50, and 65°C storage temperatures increased in length as a result of self-irradiation follows exponential dependence on dose or time during initial stage of aging. After nine equivalent years of aging, the samples at 35°C have expanded in volume by 0.08% and exhibit a near linear volume expansion behavior by helium in-growth mechanism.

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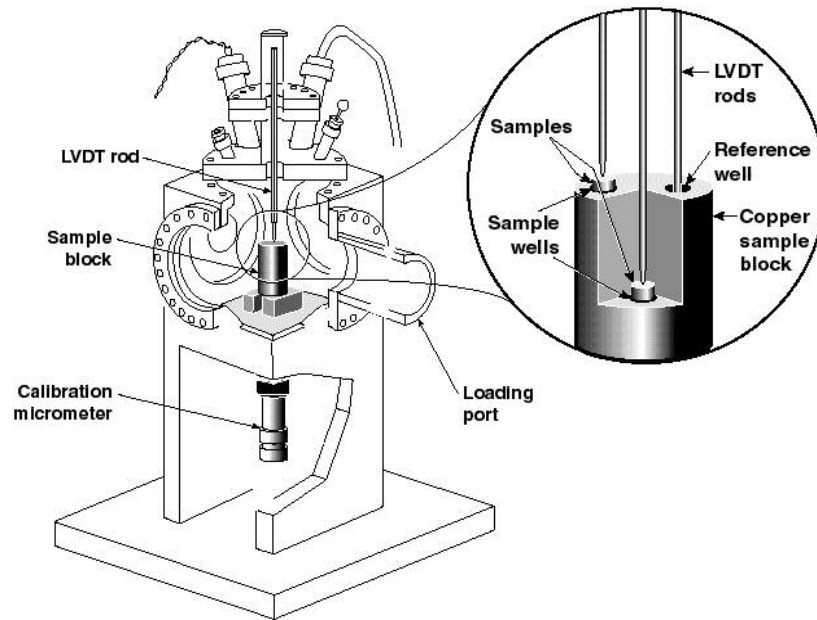


Figure 1. Design of the dilatometry system to monitor length change in Pu alloy specimens.

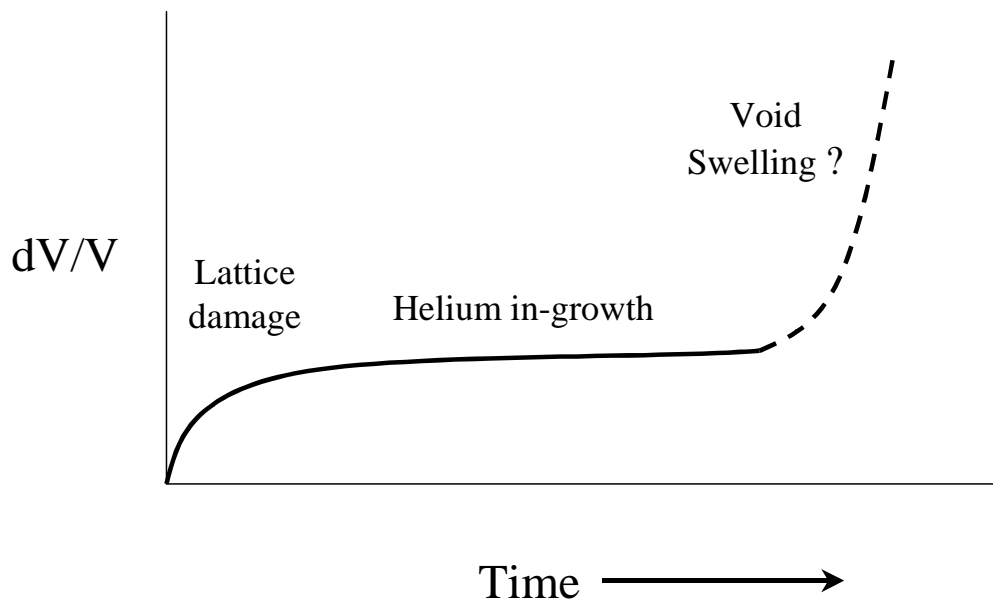


Figure 2. A possible volume change behavior of plutonium as a function of time. The dV/V represents the volume change normalized to the initial volume.